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Abstract

Using computational alchemy, the authors demonstrate how catalyst discovery can be made faster and readily accessible. Starting from electronic structure results on one system, they give predictions for barriers and binding energies for variants of that system. This cashes in on the promise that computational alchemy allows for systematical screening of chemical spaces in pursuit of a design application. Their work is written in an hands-on explicit style including clear figures which render the work accessible to the broad audience for which it is relevant.

I suggest to **accept** the article after minor revisions as outlined below.

1 Referee Report

My main question is whether the authors have seen any kind of finite-size effect on the alchemical potential due to the minimal cell employed in this study. Due to the coulombic form of the alchemical potential, it appears unlikely that there is no such effect, but potentially it is a quite systematic contribution for all target systems considered.

The authors state “Computational alchemy is a perturbation theory approximation”. It might be warranted to rather emphasise in the article that it is the Taylor series truncation that renders it an approximation. Under the supposition that the Taylor series converges, in fact the expansion should be exact. Admittedly, not all of the terms (e.g. basis function changes in the direction of the alchemical change) are typically taken into account explicitly, but only if the perturbation approach is exact in the first place, computational alchemy can expect to have the predictive power that is required for the materials design applications outlined in this work.

Similarly, when the authors discuss the charge neutrality of the alchemical change, it might be important to highlight that the theory does not require the changes in nuclear charges to be strictly compensated. In this particular application however, the change should be neutral as otherwise the total surface charge density of the interface becomes unphysically large.

When discussing the energy barriers relative to DFT data, the authors give an error bound of 0.3eV. To set this into perspective: how accurate can the DFT method expected to be for the systems at hand? Effectively, computational alchemy constitutes a Taylor expansion on the potential energy surface of the reference method used, therefore can only hope to recover the accuracy of the reference method. If this error bound is low compared to DFT vs higher level methods, this would effectively render computational alchemy to have predictive power indistinguishable from the reference method.

In Figure 5, the authors show the NEB paths found from computational alchemy. For the case of Delta Z=-1, NEB image 7 has a substantially lower energy, even lower than the endpoint of the NEB path. What is the reason for this given that alchemical predictions are done on all NEB images in the same fashion?

Figure 5 b) is hard to assess visually, since most of the data points are obscured by others. Perhaps a plot of signed difference alchemy-DFT vs distance from the transmutation site would be clearer. Moreover, no units are given.

In the caption of Figure 2, some cross-references seem to be mixed up.

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